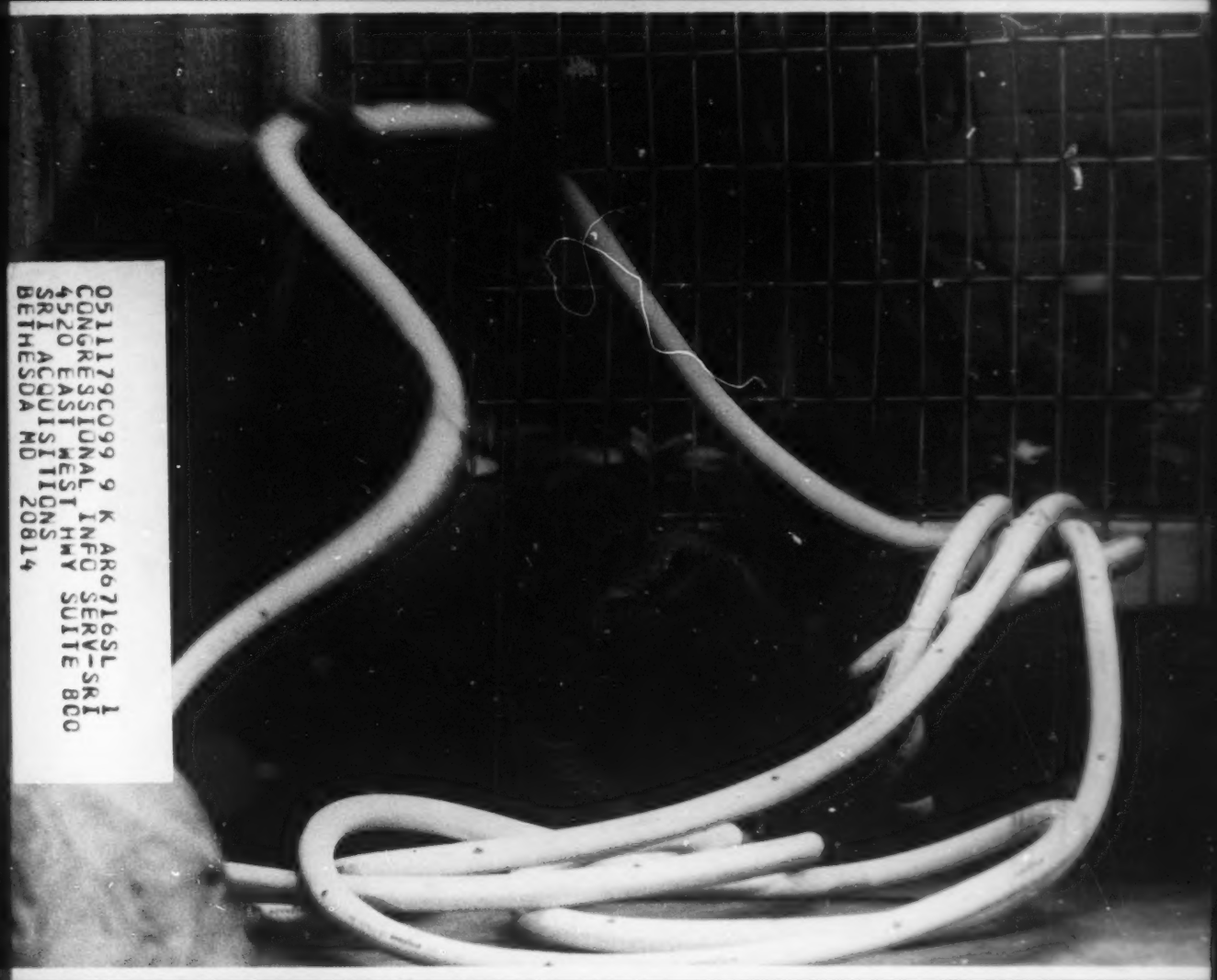
APRIL 17, 1989

CHEMICAL & ENGINEERING

NEWS



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RUBBER CHEMICALS

Producers strive to meet tougher performance demands Page 25

CHEMICAL & ENGINEERING
NEWS

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APRIL 17, 1989

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■ **Robert Maxwell, British media baron, signs protocol for joint science database development with Soviet technical information institute.** Page 8

Cover: Zoo gorilla plays with Goodyear's multipurpose rubber hose, tradenamed Gorilla for its strength, which was designed for agriculture, mining, and construction industries

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New technique, pulse oriented gel electrophoresis, produces first images of individual DNA molecules. **Page 23**

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Chemical & Engineering News

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LETTERS

Radon monitoring

SIR: I would like to commend David Hanson (C&EN, Feb. 6, page 7) on his fine, comprehensive presentation of the radon monitoring/control issue. A major point of new information was not mentioned by Hanson, however. Title III—Indoor Radon Abatement, an Amendment of the Toxic Substances Control Act was passed by both houses of Congress and signed by President Reagan in October 1988. "The bill now sets as a long-term national goal the reduction of radon in buildings to the level of the outside air" or to "background levels of 0.2 picocuries per liter." (*Congressional Record-House*, Oct. 5, 1988, pages H9637 and H9640).

One of the main purposes of the legislation is to supply individual states with money to begin extensive programs of radon testing, and to train radon testing and mitigation personnel. This amendment has significant impact on mitigation, perceived health effects, and screening measurements of indoor radon.

Mitigation down to 4 pCi per L (the present Environmental Protection Agency guideline) is well within the capabilities of current contractors and techniques. Mitigation to ambient values may or may not be possible using existing technology. Mitigation of radon in houses presently costs about $300 to $5000 for various engineering modifications; decreasing radon to more than an order of magnitude less than 4 pCi per L may cost up to an order of magnitude more.

Potential deleterious health effects of radon have not been well documented experimentally below 4 pCi per L. Further research will be necessary to ascertain the true relationship of low concentrations of radon in air to cancer in humans.

Radon levels in indoor air fluctuate drastically with changes in soil moisture, barometric pressure, outside wind movement, outdoor/indoor temperature differences (the stack effect), and different building conditions during testing. Therefore, defining the true annual average concentrations in buildings may be difficult. The most commonly used inexpensive measurement techniques (charcoal cannister; track-etch) suffer from uncertainties of up to ±25% (two standard deviations, at the 90% confidence level) for side-by-side duplicate analyses, and uncertainties up to ±50% for track-etch to ±90% for charcoal cannisters (90% confidence level) in projecting annual exposures from a single measurement (personal communication from D. G. Mose, George Mason University). Therefore, defining the true annual radon concentrations in buildings will be difficult and expensive under the new guideline.

The amendment strongly recommends (but does not mandate) as a national goal mitigation of all homes and buildings to the lowest feasible level below 4 pCi per L. EPA has been directed to aggressively promulgate the newly recommended ambient indoor radon guideline and emphasize the hazard of radon concentrations well below the 4 pCi per L former guideline. However, such federal guidelines often carry the weight of law. As tenants sue building owners over elevated radon exposure, state-of-the-art and best standard practices arguments in civil suits may eventually require building owners to mitigate radon to ambient concentrations.

E. Kent Sprague
McCrone Environmental Services Inc.
Norcross, Ga.

Fluoridation of water

Sir: The letter by Surgeon General C. Everett Koop on fluoridation of water (C&EN, Nov. 28, 1988, page 2) and subsequent letters to the editor illustrate the apparent flexibility of the scientific method in connection with a political goal. The letters accurately point out the need for more stringent criteria, more open discussion among scientists concerning the quality of the scientific data, as well as the limitations of the scientific method before conclusions reach the popular media and political leaders. Perhaps the best example of public policy built on a foundation of emotion and political goals rather than rigorous science is that of environmental tobacco smoke and the alleged health effects on the nonsmoker. Since the Surgeon General's report of the health consequences of involuntary smoking, which concluded that "involuntary smoking is a cause of disease including lung cancer in health of nonsmokers" (which was not adequately supported by the evidence presented in the body of the report), the statistically estimated death toll attributed to environmental tobacco smoke set forth in published reports has escalated from 5000 [*Environ. Int.*, **11,** 3 (1985)] to 46,000 [*Environ. Int.*, **14,** 249 (1988)]. Although these reports were peer reviewed, they suffer from many of the limitations cited in the article by Alvan R. Feinstein in *Science* [**242,** 1257 (1988)] regarding the need for improved methods in epidemiology.

The unsubstantiated number of

Employee at Goodyear's radial passenger car tire plant in Union City, Tenn., checks high-performance tire on production line

Rubber Chemicals Face More Demanding Market

Bruce F. Greek, C&EN Houston

Demand for rubber-processing chemicals has surged during the past two years. The upswing resulted from a global rise in demand for various kinds of tires and fabricated rubber products. Future growth for these chemicals will follow growth of rubber products use because unit consumption of chemicals in rubber is expected to change very little during the next few years.

Rubber-processing chemicals play a key role in the manufacture of rubber products because they provide desirable properties to commercial rubber products. They aid in rubber vulcanization, protect polymer chains, and make processing of elastomers easier and less costly. Seldom is a rubber product fabricated without the help of such chemicals.

Output of U.S. rubber-processing chemicals this year is expected to top 400 million lb. Production in 1988 was close to 400 million lb. Although expected to increase, output of rubber-processing chemicals won't grow so much as in 1987 nor so much as in 1986 when the current boom in rubber chemicals began.

Capacity to make these chemicals is operating full out. Minor debottlenecking and process-efficiency improvements are the sources of any increases in capacity; no new plants or units have been announced this year.

Rubber-processing chemicals usually are defined as those that react with the rubber. Some may be part of the vulcanization process, and are known as accelerators or retarders because of the way they affect that process. Others, called antidegradants or age resisters, decompose to prevent a degradative reaction of elastomers with oxygen or to minimize effects of light and heat on polymers. Yet other chemicals are used to break polymer chains, making processing quicker and easier and otherwise aiding the fabrication process.

Some chemicals that rarely react with the elastomer are used in compounding rubber for fabrication.

Rubber-processing chemicals at a glance

Most finished rubber products consist of elastomers and a host of additives. The additives are used to provide the best possible cost/performance for the products. Many complex organic chemicals—rubber-processing chemicals—aid the vulcanization process; reduce attack on the elastomer polymer by a combination of oxygen, ozone, and light; and make the fabrication of rubber products easier

U.S. production: (1987) 382 million lb

U.S. merchant sales: (1987) quantity—289 million lb, value—$359 million

Producers: More than 20 in U.S.

Suppliers: More than 50 in U.S. of formulated products for rubber compounding

Compounds: Several hundred compounds are known to have use in rubber processing, but fewer than 200 are of commercial significance. Examples of significant compound groups: benzothiozoles, *p*-phenylenediamines, phosphites, polyphenols, and dithiocarbamic acid derivatives

In the rubber industry, however, they are considered to be outside the group known as rubber-processing chemicals. Some examples are carbon black, titanium dioxide, silica, and waxes. Carbon blacks and many inorganic materials are used as reinforcements, fillers, and colorants.

Sulfur is especially important to the vulcanization process, but it, too, generally is excluded from the group referred to as rubber-processing chemicals. Other chemicals also are used in different kinds of vulcanization or curing processes. Large quantities of fabrics made of synthetic or natural fibers, glass, and metals also may be used in fabricated rubber products.

In 1987, more than 20 companies produced a total of 382 million lb of rubber-processing chemicals, according to the U.S. International Trade Commission (ITC). Output was up 58 million lb, or 18% from 324 million lb in 1986. The ITC data are questionable, according to several company officials, who point out that many rubber-processing chemicals have applications in other industries. That situation leads to potentially subjective allocation by end-uses and to the high growth rates. Producers consequently put production growth of rubber-processing chemicals between 4 and 7% annually for 1986 and 1987.

Although U.S. demand for rubber has increased modestly during the past two years, world demand has increased more rapidly, causing the consumption of rubber-processing chemicals to rise globally. The rise in global demand has spurred exports of U.S. rubber-processing chemicals. Until production became limited by available capacity, U.S. exports also were spurred by the decline in the value of the dollar relative to many other currencies. Lack of expected sales of rubber-processing chemicals produced in East bloc countries, despite construction of new plants in these countries to make these chemicals, has been another factor in rising exports. Expansion of demand for rubber products in East bloc countries apparently has absorbed output from the new capacity there.

Nearly all plants making rubber-processing chemicals in noncommunist countries currently are operating at capacity. The few expansions of capacity now under way are minor, although they are larger than debottlenecking projects. Thus tight supply, limited increases in production, and some additional selling price increases will make up the business during 1989, say some industry sources. Trade in these chemicals will increase faster than production because of efforts of consumers in various parts of the world to get supplies of chemicals they need. No real shortage of acceptable chemicals is expected, however, because capacity to fabricate rubber products also is running nearly full out.

Additional capacity to make rubber-processing chemicals likely will be needed later on. Despite possibilities that economic slowdowns will occur, significant increases in rubber consumption are forecast. For example, consumption of both synthetic and natural rubber in noncommunist countries will increase 11% between 1988 and 1993, reaching 12.1 million metric tons, according to the latest study of the International Institute of Synthetic Rubber Producers (IISRP). Consumption of synthetic rubber will grow faster than that of natural rubber. Demand for most types of synthetic rubber will increase during the next five years, with that for ethylene-propylene-diene rubbers predicted to have the largest growth.

Annual consumption of all kinds of rubber will grow slowly, suggesting that small debottlenecking

Production of rubber-processing chemicals on the upswing

	CYCLICS								ACYCLICS			
	Accelerators[a]			Antidegradants[b]								
Millions of lb	Thiazoles	Others	Total	Amine compounds	Phenolics and phosphites	Total	All others	Total cyclics	Accelerators[a]	Others	Total acyclics	Grand total
1983	62.9	5.1	68.0	105.6	63.4	169.0	9.1	246.1	8.8	37.7	46.5	292.5
1984	80.3	9.3	89.6	98.0	62.7	160.7	9.5	259.8	8.8	19.0	27.8	287.6
1985	63.9	5.2	69.1	100.2	59.3	159.5	8.6	237.2	6.9	16.1	23.0	260.2
1986	52.1	4.3	56.4	112.6	67.8	181.3	59.1	296.8	na	na	27.3	324.1
1987	80.9	4.2	85.1	123.2	80.5	203.7	65.6	354.4	5.3	22.4	27.7	382.0

a Includes activators and vulcanizing agents. b Includes antioxidants, antiozonants, and stabilizers. na = not available. Source: International Trade Commission

Special chemicals are essential to rubber compounding

Small amounts of rubber-processing chemicals provide large benefits in physical properties of finished rubber products. Rubber-processing chemicals strongly affect the degree and time of the vulcanization of rubbers; influence greatly the flexibility of the final fabricated products; help elastomers resist breaking up from attack by oxygen, ozone, light, and heat; and reduce the harmful effects of hysteresis or heat buildup. They are essential to providing rubber goods that perform well at low cost.

Rubber-processing chemicals of greatest commercial value are those used in rubber processing prior to fabrication and also during fabrication itself to provide many desirable properties. Other materials used in compounding rubber for subsequent fabrication are vulcanizing agents, most of which include a sulfur atom or are peroxides, metallic oxides, or urethane-type compounds; carbon blacks (particularly used to give abrasion resistance, tensile strength, and tear resistance); and fillers such as clays, silicas, and calcium carbonate. Other chemicals involved in elastomer production are catalysts for polymerization, controls for molecular weight, and antioxidants to stabilize raw polymers.

Some rubber chemicals are multifunctional in providing desired properties to the fabricated rubber products and in aiding the rubber compounding process. However, most have a single use. Often more than one compound may be used to cause an action during compounding or curing of rubber or to give a specific property to a final product.

Not all functions of rubber chemicals are required in all fabricated forms, but the major functional types are added to most compounds. The quantities used may vary quite widely, depending on the desired performance of the products. The commercially significant functions of rubber chemicals are:

Accelerators. Many different compounds are used to speed, to varying degrees, the vulcanization of rubber. Without accelerators, vulcanization of rubber would be so slow that fabricated products would be far more costly.

Activators and retarders. These chemicals increase or decrease the effects of accelerators. Activators are used far more than are retarders. Most activators are alkaline materials. Conversely, most retarders are acidic. Retarders are used much less often and mainly to minimize premature vulcanization during processing.

Antidegradants. Sometimes called age resisters, this very large class of compounds includes antioxidants, antiozonants, ultraviolet light protectors, and antiflex agents. Most of these compounds are designed to work by inhibiting the reactions between oxygen and elastomers. Carbon blacks and other materials used in compounding rubber also provide protection against degradation. However, because those materials have other primary uses, they are usually classified elsewhere.

Miscellaneous rubber-processing chemicals. This group includes the many processing aids used to reduce the time and energy required to prepare elastomers for the fabricated products. It also includes bonding agents, dispersing agents, colorants, flame retardants, and blowing agents. Peptizers, many of which are organic sulfur compounds, help to break polymer chains, and are the most commercially significant of the group. Other processing aids are various synthetic resins, fatty acids and derivatives, petroleum fractions in the lube oil range of hydrocarbons, and various natural fats, oils, and pine products. A host of bonding agents, many of them proprietary materials, are used to bind the elastomer to metals and textiles that make up the composites that are fabricated rubber products.

and efficiency improvements in plants that make processing chemicals might provide some capacity increases to meet part of expected added demand. For example, demand for the synthetic rubber produced in largest volume, solid styrene-butadiene rubber (SBR), amounted to 2.38 million metric tons in 1988 in noncommunist countries, according to IISRP estimates. This year use is forecast to rise to 2.41 million metric tons, and to increase 8.8% to reach 2.62 million tons in 1993. Demand for SBR in latex forms is forecast to rise from 1.34 million metric tons this year to 1.45 million tons in 1993.

IISRP forecasts demand for other synthetic rubbers to grow by similar rates. Polybutadiene demand also will increase 8.8% from 1.12 million metric tons in 1989 to 1.22 million tons in 1993. The growth in demand for ethylene-propylene rubbers will be higher, at more than 15%, over the five-year period. Demand for other synthetic rubbers, including butyl and polychloroprene, will climb 123,000 metric tons by 1993, or 12% over the 1.04 million metric tons forecast for this year.

Demand for natural rubber in noncommunist countries will slightly exceed that for synthetic rubber. Between 1989 and 1993, use of natural rubber will rise 9.8%, IISRP forecasts. The area with the fastest growing demand for natural rubber is Asia and Oceania, where total rubber use in 1993 will exceed that of Canada and the U.S.

If demand for fabricated rubber products follows current forecasts, construction of new capacity for rubber-processing chemicals will begin too late to stave off a further tightening of supplies. No large, new units are seriously planned in the U.S., in Western Europe, or in the Far East. Competition during weak periods of demand for rubber-processing chemicals during the early 1980s pushed down prices. By 1986, the average unit price for U.S. merchant sales had declined about 25%, according to ITC. During the latter part of 1987, however, prices for most rubber-processing chemicals began to rise, along with prices of other chemicals. Producers say that, though prices continue to rise, they generally have not returned to levels of a decade ago. Meanwhile, production costs have risen substantially. Therefore, returns remain well below reinvestment levels, especially since costs of plant construction started to rise again about the same

time as did selling prices for rubber-processing chemicals.

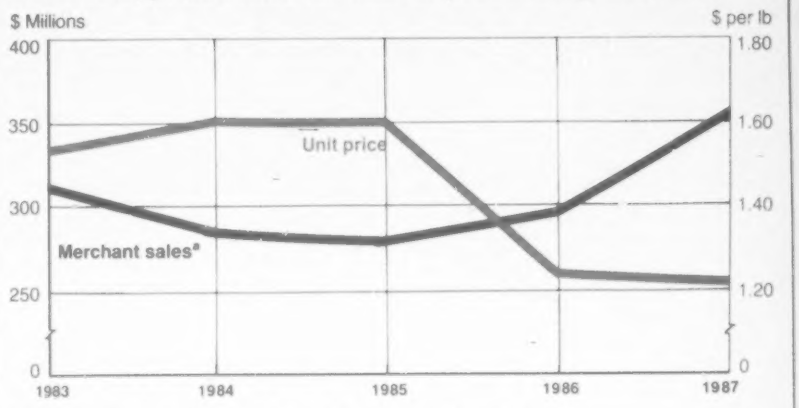
Tight supplies and higher prices for rubber-processing chemicals suggest a shift toward lower use. Fabricators also seek other ways to provide the desired properties in rubber products. Despite research on ways to reduce use of rubber-processing chemicals (begun just after vulcanization was discovered), their use has slowly increased—although growth has been limited by discoveries of new chemicals that give better properties than those obtained with currently used chemicals. Because of cost/performance requirements, rubber-processing chemicals currently are used at levels sufficient to give desired properties without the need of surpluses to cover unexpected conditions.

Because of changes in fabricated product demand, kinds of elastomers used in products, and fabrication technology, some changes are expected in consumption of these chemicals. For example, changes made in elastomers and improvements in compounding will further counteract effects of oxidation and ozone. Thus, lower antidegradant use will be required, according to results of a study by Freedonia Group, a Cleveland consulting firm. According to Freedonia, however, more accelerators and activators will be needed because of efforts to improve rubber-processing and fabrication efficiencies. The firm projects that consumption of processing chemicals on a unit basis will rise to 133 lb per ton of rubber consumed in 1991 and to 140 lb per ton in 1995 from 122 lb per ton estimated to have been consumed in 1986. Coupled with the small rise in unit consumption, higher selling prices will raise the value of U.S. consumption of rubber-processing chemicals at an average annual rate of 5.7% between 1986 and 1991.

Tires and related products account for more than 60% of new rubber consumption in the U.S. annually. A larger share of natural rubber consumption goes into tires compared with the share of synthetic rubber used in tires. For example, IISRP forecasts that this year tires and tire products will account for 53% of U.S. synthetic rubber consumption, excluding carboxylated styrene-butadiene latex and nitrile latex. Tires and tire products consumption of synthetic rubber will rise, but not so fast as consumption by other rubber products, which will result in the decline in share of consumption by tires and tire products.

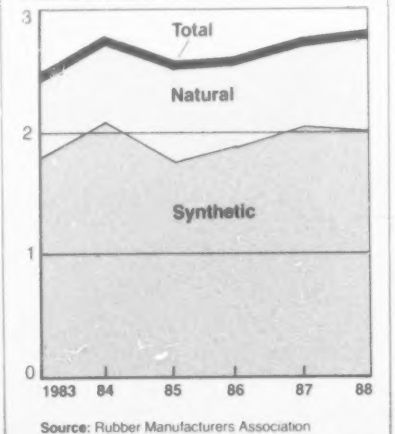
Production of passenger car, truck, and bus tires in 1988 totaled 211 million units, according to the Rubber Manufacturers Association. Additional quantities of aircraft, tractor and farm implement, and special industrial tires also were produced. The excess of imports of tires over exports added more than 28 million passenger car, truck, and bus tires to the U.S. supply.

Sales rise, prices hold for rubber-processing chemicals



a Excludes chemicals consumed captively. **Source:** International Trade Commission

U.S. rubber consumption continues to increase



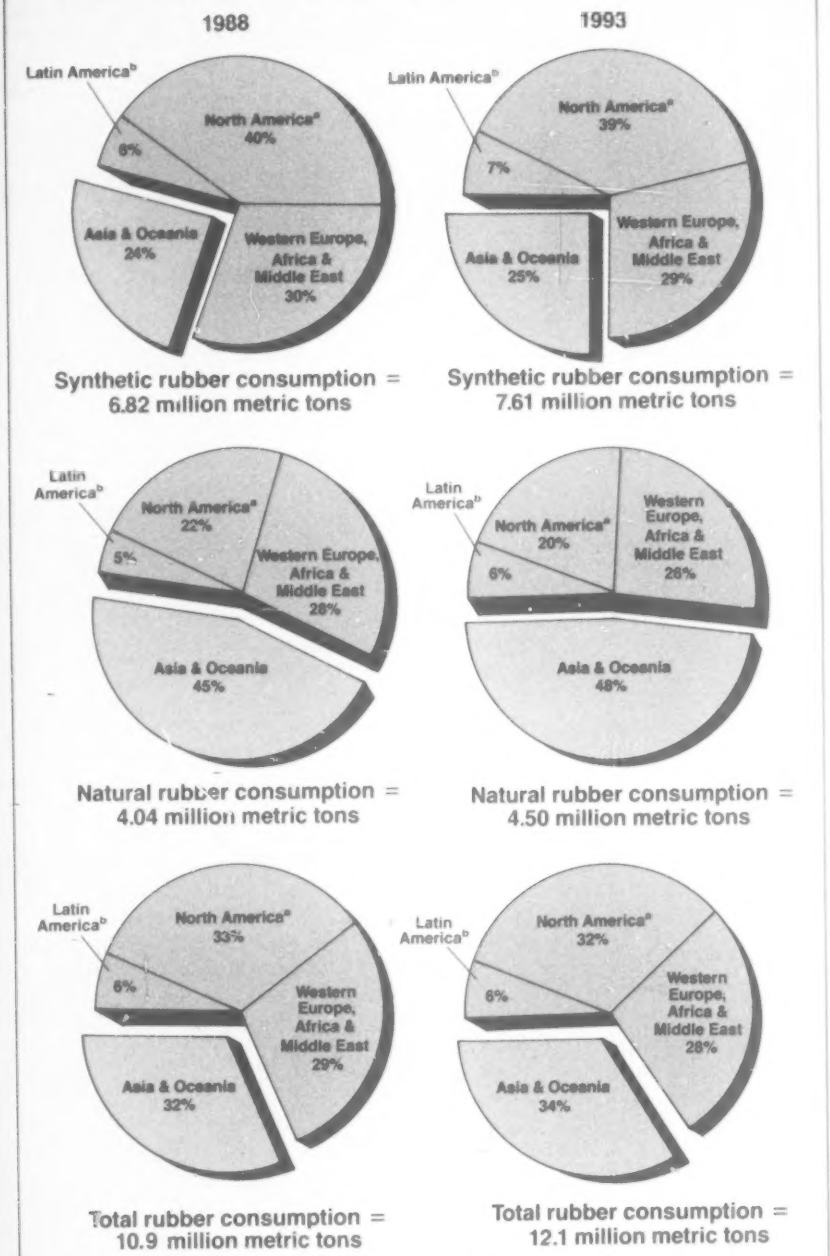
Source: Rubber Manufacturers Association

U.S. passenger car, truck, and bus tire output has held at 200 million units, plus or minus 5% annually for five years. Until some of the capacity expansions that are under way come on stream, tire plants will continue to be utilized at capacity, which tends to hold down production. A large quantity of tire-building capacity was scrapped during the early 1980s when the shift from bias-ply to radial tires for passenger cars neared completion and tire-tread life increases reduced replacement demand. During that time, imports of tires increased rapidly, as did imports of autos, many of which carried tires produced outside the U.S.

Some $2 billion will be spent on tire-building capacity in the U.S. by 1992, if present plans go ahead. In addition, another $4 billion is forecast to be spent for tire capacity in the rest of the noncommunist world. How much these expenditures will increase tire-building capacity is uncertain. A large part of the outlay is going to plants that will build radial truck and bus tires. The conversion from bias-ply truck and bus tires is now well under way. Another part of the expenditures will go to modernizing production facilities in an effort to reduce manufacturing costs.

Changes in tire demand is another uncertainty. For most U.S. cars, and a significant segment of trucks and buses now operating on radial tires, average tire life is growing

Asia and Oceania will become largest rubber-consuming region in Western world by 1993


1988
1993
Latin America[b]
North America[a] 40%
6%
Asia & Oceania 24%
Western Europe, Africa & Middle East 30%
Synthetic rubber consumption = 6.82 million metric tons
Latin America[b]
North America[a] 39%
7%
Western Europe, Africa & Middle East 29%
Asia & Oceania 25%
Synthetic rubber consumption = 7.61 million metric tons
Latin America[b]
North America[a] 22%
5%
Western Europe, Africa & Middle East 28%
Asia & Oceania 45%
Natural rubber consumption = 4.04 million metric tons
Latin America[b]
North America[a] 20%
6%
Western Europe, Africa & Middle East 26%
Asia & Oceania 48%
Natural rubber consumption = 4.50 million metric tons
Latin America[b]
North America[a] 33%
6%
Western Europe, Africa & Middle East 29%
Asia & Oceania 32%
Total rubber consumption = 10.9 million metric tons
Latin America[b]
North America[a] 32%
6%
Western Europe, Africa & Middle East 28%
Asia & Oceania 34%
Total rubber consumption = 12.1 million metric tons

a Canada and U.S. b Includes Mexico. **Source:** International Institute of Synthetic Rubber Producers

more slowly than five years ago. A recent shift to higher use of high-performance and off-the-road tires on passenger vehicles offsets the impact of radial tires upon increasing tire life. These high-performance and off-the-road tires, because of their design and use, have shorter tread life than do conventional radial passenger car tires. Although demand will not be noticeably affected by such changes in tire type and use for several years, additional tire-building capacity may be necessary in the 1990s.

Because of the importance of the tire industry as a consumer of rubber, tires will continue to dominate the markets for rubber-processing chemicals. Tire use of chemicals continues to grow, but not so fast as in mechanical goods and other rubber products excluding footwear. Hence, consumption of chemicals in tires is slowly losing share of the market. The tire market, nevertheless, continues to spur developmental efforts by producers of rubber-processing chemicals. Chemicals used in tire fabrication generally are tested for possible use in fabrication of other rubber products, and they are used where possible. Higher value uses in mechanical goods for specific properties will lead to development of a chemical for such uses, but that work is small when compared with chemical development for tire consumption.

Any growth in tire production, therefore, may be expected to drive growth in rubber-processing chemicals. If tire production should increase rapidly as a result of sizable additions in U.S. and foreign capacity, two concerns arise. One pertains to where the additional tires will be sold and suggests a resurgence of competitive pressures. Part of the new capacity will be used for tires going to autos and trucks to be sold in the faster growing markets of developing countries. The remainder of capacity would go to replacing obsolete facilities in the U.S., Western Europe, and Japan, or would be used in competition for market share in existing markets.

The second concern is the availability of capacity for rubber-processing chemicals used in making the tires. Chemical producers, only now seeing recovery in prices after declines in the early 1980s, are expected to be wary of making large investments in plants if selling prices of their products decline under pressure from tire makers who face price competition in tires they sell. Because of the complexity of manufacturing rubber-processing chemicals, sizable new entrants into the market are unlikely.

However, new entrants into rubber-processing-chemicals markets would also be expected to provide the services that producers developed in the 1980s to sell their chemicals in response to competitive pres-

sures. Offering the high degree of technical service and product quality that now is expected of companies to give product differentiation is another barrier to potential new suppliers. Small companies can carve out a niche, but their contribution to the volume needed would do little to increase world supply.

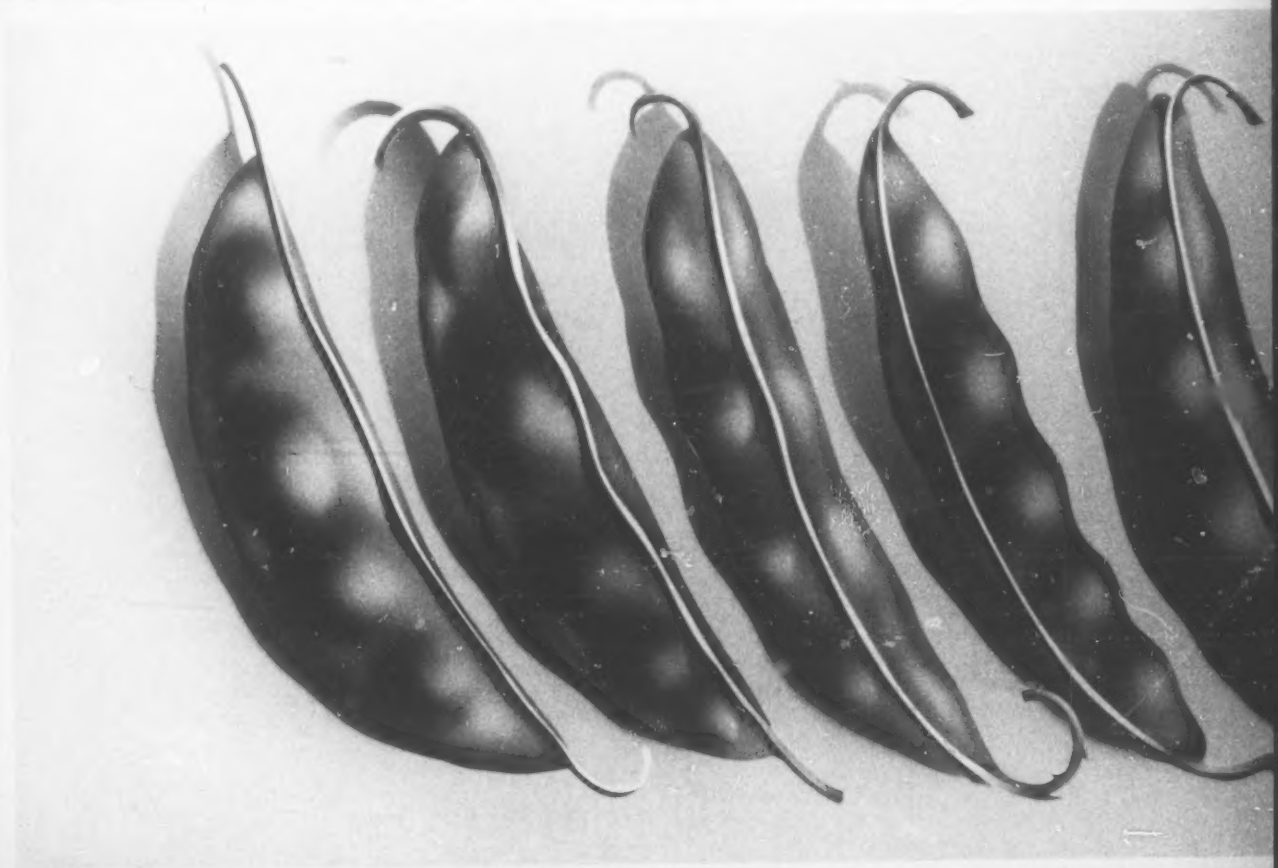
With the changes expected in tire capacity, efficiency of tire building has become more important. Hence, rubber-processing chemicals are needed to improve the efficiency, with much emphasis on the vulcanization process. Except for butyl rubber and some rubbers produced in small quantities, large-volume rubbers can be cured with peroxides. However, most vulcanization still is done by using sulfur or a sulfur-donating chemical. The sulfur vulcanization process causes polymer chains to be connected through sulfur atoms into a three-dimensional form, which provides much greater elasticity and less plasticity. Controlling not only the quantity of materials added to the rubber but also the heat and time influences the properties of the vulcanizate.

Early rubber processers found that use of sulfur and inorganic salts alone took a long time and produced a rubber with poor properties. Minor improvements were made over 50 years, but development of a practical car in about 1900 required pneumatic tires and rubber that was better compounded. Organic chemicals, starting with aniline, were found to speed greatly the vulcanization process and to produce fabricated rubber products with vastly better properties. Toxicity of these chemicals was recognized soon after initial use, and efforts were made to find even better and safer chemicals as vulcanization aids.

The vulcanization aids soon were called accelerators because that was what the first ones did for the vulcanization process. From first use of aniline, a whole business of rubber accelerators, activators for accelerators, and the reverse—retarders—has developed. Products of reactions with aniline aimed at reducing toxicity developed. Some were found also to cause improvement in aging characteristics of rubber. The second major field of rubber-processing chemicals—the antidegradants—then developed.

Numerous organic chemicals—most having one or more sulfur or nitrogen atoms—were tested as vulcanization accelerators prior to the mid-1920s. Among them: dithiocarbamates, xanthates, guanidines, and thiurams. However, nearly all of the compounds in these groups had deficiencies. They frequently were unstable during processing or were overactive and less controllable than processors wished. Some of these accelerators are still used today in limited quantities for specialized types of fabricated rubber products.

The search for better accelerators



has led to 2-mercaptobenzothiazole (MBT) and to development of a whole class of benzothiazole accelerators. Of organic accelerators, activators, and vulcanizing agents reported to ITC, the thiazoles account for more than 90% of production. It is estimated that today more than 100 different compounds, blends, and proprietary mixtures can be obtained commercially for use as accelerators.

The advent of synthetic rubbers and new types of carbon black with finer particle sizes has spurred the search for better accelerators. But compounds of rubber, carbon blacks, and fillers have become more difficult to process. Temperatures during the mixing rise, possibly causing vulcanization or curing to occur sooner than desired. Rubber in which premature vulcanization occurs will have poor properties. Such rubber is called scorched, and is likely discarded or used in a low-value product.

The tendency for MBT accelerators to scorch synthetic rubbers prompted Monsanto to develop a class of compounds, sulfenamides, made by reacting MBT with various kinds of amines. Sulfenamides, also now sold by several rubber-processing chemical suppliers, account for a large share of the chemicals sold as accelerators. They are sometimes called delayed-action accelerators. Cure rates and scorch safety (time before scorch occurs) will vary, depending on selection of the amine used in making the benzothiazolesulfenamide. Highly hindered amines or less basic amines generally will cause slower cure rates.

Some thiazoles, other than the sulfenamide derivatives, are being replaced in rubber fabrication because of growing recognition that decomposition products may include nitrosamines, which are suspected carcinogens. The nitrosamines have been detected in concentrations higher than normal in locations where rubber products are made or stored. They are being brought under toxic substances control legislation that now is either in place or is being considered for enactment in many countries, including the U.S.

The smaller volume commercial accelerators include dithiocarbamates and derivatives such as the thiurams. In the rubber industry, these compounds are considered as ultra-accelerators because they have fast curing abilities. In some vulcanization processes, these compounds will provide enough internal sulfur to produce vulcanizates that contain short cross-links and that have good resistance to heat and oxygen. They also may be used with thiazoles and sulfenamides in proprietary formulations so that the vulcanization rate can be adjusted. These materials most often are used in fabricating hose and other mechanical rubber goods.

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Monsanto technician tests sample of rubber product at company's Akron R&D lab

other materials known as activators frequently are used to increase the effects of accelerators in rubber fabrication. Stearic acid and zinc oxide long have been a combination that rubber processors favor, because the cross-linking is speeded up. Other fatty acids (often mixtures such as are found in vegetable oils) and other metal oxides (such as litharge or magnesia) also are used as activator systems. Despite extensive study, the way that activators work is not yet clearly known.

Neither is the process by which retarders work to slow premature vulcanization fully understood. Retarders are used to extend the time to prepare rubber products for vulcanization (including the various steps from mixing to assembling the final product). Retarders now most widely used in rubber processing are aromatic acids, anhydrides, and phthalimides, or other complex amine derivatives. Choice of retarder will depend on several factors: the kind of elastomer being processed, the accelerator or accelerator system being used, the amount of processing needed, and the temperatures to be used. Use of retarders represents a trade-off between prevention of materials loss (in prematurely vulcanized rubber compounds) and lower productivity because of longer times required during molding.

Long-term performance of the rubber product depends on its retaining good physical properties. Most elastomers are attacked rather rapidly by oxygen, ozone, heat, light, mechanical stress and fatigue, and various chemicals (all of which reduce performance of the product). Chemicals added to rubber compounds to prevent attack on the elastomers generally are called antidegradants, or age resisters. Carbon black and other materials also will significantly help compounded rubber to resist degradation. These materials, however, usually are listed separately from the group of antidegradants. Surface coatings some-

Many chemicals are used to vary vulcanization rate

	Relative rate of vulcanization	Typical compound
Aldehyde/amines	Fast	Butyraldehyde-aniline
Dithiocarbamates	Very fast	Zinc dimethyldithiocarbamate
Guanidines	Slow	1,3-Diphenylguanidine
Sulfenamides	Fast	*N*-cyclohexyl-2-benzothiazolesulfenamide
Thiazoles	Medium	2-Mercaptobenzothiazole
Thioureas	Medium	Ethylene thiourea
Thiuramsulfides	Fast	Tetramethylthiuramdisulfide

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Styrene-butadiene rubber holds share of consumption

Thousands of metric tons	1987	1988	1989[a]	1993[a]
SBR solid	2324	2382	2405	2616
SBR latex	241	245	250	265
Carboxylated latex	984	1049	1088	1189
Polybutadiene	1044	1098	1117	1215
Ethylene-propylene	498	540	557	642
Polychloroprene	249	251	251	258
Nitrile (solid and latex)	221	237	240	262
Other synthetics[b]	958	1022	1040	1163
TOTAL	6519	6824	6948	7610

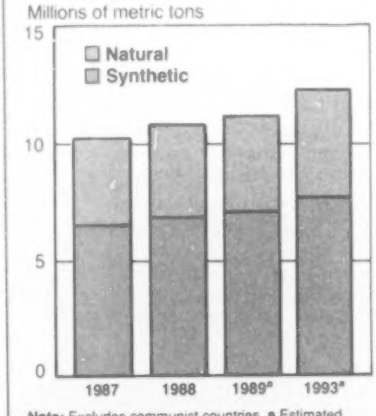
Note: Excludes communist countries. a Estimated. b Includes butyl and polyisoprene. Source: International Institute of Synthetic Rubber Producers

times applied to mechanical goods (to prevent oil or ozone from reaching the elastomer) also are excluded from the antidegradant group.

Results of the oxidation of elastomers appear in either of two ways—hardening or softening of the rubber. Cross-linking during oxidation will occur in rubbers containing butadiene molecules, increasing the rubbers' stiffness and brittleness and decreasing elongation. This will give a leatherlike feel to the surface of the rubber. If the rubber contains an isoprene molecule, softening and weakening will occur, and a stickiness will appear as the elastomer chains break.

Much of the effort toward developing antidegradants is directed at antioxidants. Studies in oxidation of elastomers and the function of antioxidants in reducing the effects of oxygen currently show the possibility that antioxidants moderate the polymer chain degradation by interfering with reactions of oxygen-containing radicals formed from the initial oxidative attack or from chain termination.

Global use of natural and synthetic rubber to rise



Note: Excludes communist countries. a Estimated.
Source: International Institute of Synthetic Rubber Producers

The performance demands that have been put on rubber products for more than 100 years have led to the testing of a wide variety of chemicals as antioxidants, many of which were then discarded. Currently, the most widely used antioxidants fall into the categories of amines, phenols, and thioesters. Diphenyl amines, paraphenyl diamines, and dihydroquinolines are the most used amine antioxidants. Various naphthylamines are nearly no longer used because of concerns over toxicity.

Most commercial antioxidants that contain a phenolic structure fall into five general classes: hindered phenols (with varying side groups on the ring), hindered bisphenols, hindered thiobisphenols (in which sulfur replaces a hydrocarbon group in the bridge structure), polyphenols, and polyhydroxyphenols. Some of the phenolic antioxidants now are used more widely to stabilize synthetic rubbers prior to compounding than they are to provide antioxidant properties to the finished product.

Phosphite antioxidants frequently are used as elastomer polymer stabilizers, and probably more go to this use than to compounding. Many chemical structures of phosphites are available from rubber-processing chemical suppliers, including both alkyl and aryl groups designed for specific applications. These materials often are sold as proprietary mixtures that also may contain other processing chemicals.

Industrial work boots use variety of rubber-processing chemicals

Many of the amino compounds used as antioxidants provide protection against ozone. *p*-Phenylenediamines have been and continue to be the most widely used class of antiozonants in rubber products. Many different derivatives of these diamines are relatively easy to make by reacting various ketones and *p*-phenylenediamine. Proprietary compounds sometimes are sold for specific elastomers or for use in producing certain kinds of fabricated rubber products. Other lesser used antiozonants include such groups of compounds as dihydroquinolines, thioureas, and dithiocarbamates.

For protection against attack by ozone, paraffin wax likely is used in greater quantities than synthetic chemical antiozonants. Wax usually is blended into the rubber compound and dissolves into it during mixing if temperatures are high enough. Otherwise, it will dissolve later during the heating to produce vulcanization. Because wax has limited solubility, it will "precipitate" when the rubber product cools and will migrate to the surface of the product. At the surface, it provides a layer of material attacked very slowly by ozone. If the wax on the surface is removed by abrasion or dynamic stress, the rubber will be attacked by ozone until more wax migrates to the surface of the rubber product. Because scuffing or flexing tends to remove waxes, chemical antiozonants also are used in most rubber products where any movement at all is anticipated.

Selecting the wax to be used has become as complex a matter as selection of the other chemicals used in rubber processing. There are wide variations in the kinds of waxes available to rubber compounders. Low-melting-point waxes (extracted from cuts of crude oil with boiling points in the range of lubricating oil stocks) largely are straight-chain paraffins with low molecular weights. Rubber compounders include some of the high-molecular-weight and branched-chain paraffins (such as microcrystalline waxes) to give long-term protection against ozone.

The diffusion rate of waxes added to rubber compounds for ozone protection is affected by other components of the compound (such as type and quantities of fillers) and by temperature. At low temperatures, the low-molecular-weight waxes will migrate to the surface of the rubber product fast enough to give protection. At higher temperatures, the low-molecular-weight waxes remain soluble in the rubber and don't migrate. To get ozone resistance, high-molecular-weight waxes must be included in the wax package that is added to the rubber compound.

Adding a relatively large quantity of an ozone-resistant rubber [such as terpolymers of ethylene, propylene, and diene monomers (EPDM)] to the compound is another way to protect elastomers such as styrene-butadiene rubber and natural rubber. Although this technique will work well for some products, it may change the properties of the final product in ways unacceptable for marketing in others. One exam-

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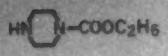
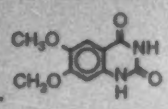
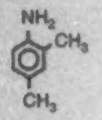


1-Ethoxycarbonylpiperazine



6,7-Dimethoxy-quinazoline-2,4-dione

2,4-Xylidine and all other isomers



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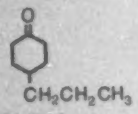
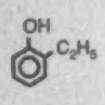
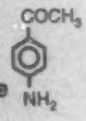


4-Propylcyclohexanone



2-Ethylphenol



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ple of attempting to use EPDM in this way is in tires, which can be severely attacked by ozone in the sidewalls if antiozonants are not added to the sidewall compound. So far, EPDM in sidewalls (other than in the decorative white stripe) hasn't given the flexing properties that are desired for long tire life.

Combining well the different types of elastomers, accelerators, activators, antidegradants, the several fillers and reinforcers, and other materials useful for fabricating a rubber product requires efficient mastication. Such mastication of rubber is necessary to make it later form smoothly. Elastomers, whether produced from tree latex or synthetically, generally are tough. They need materials that will reduce the very high viscosity to allow easier mixing of all the other components that make up a rubber compound for use in fabricated products. These materials also will lower processing temperatures that otherwise could cause scorching, and they will reduce the energy consumption required for mixing.

A large number of different materials—called peptizers as a group—have been and are being used in mechanical processing. New peptizers come to market frequently with better properties than those of currently used peptizers. As a result, numerous changes are made in the materials used in fabrication plants. Peptizers act on elastomers to reduce molecular weights of the polymers by chain cleavage and viscosity of the polymers. Use of peptizers rarely will affect vulcanization or final properties of the rubber product.

Many of the peptizers are complex aromatic disulfides and mercaptans that work by initiating radical sites that cleave polymer chains. They also neutralize previously formed radicals and promote oxidation. Natural rubber peptization is done most easily, and peptization of nitrile rubber is the most difficult among the major volume elastomers.

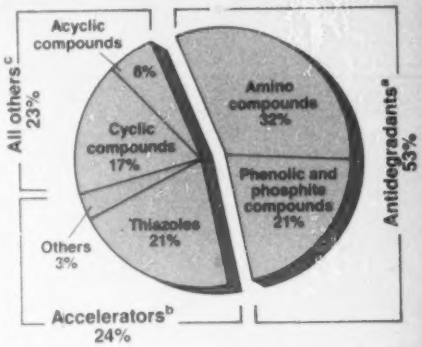
Semipeptizers is a name given to a group of sulfonated compounds and mineral oils used to break down elastomers. These materials appear to work mechanically to break polymer chains during mastication. Because costs are lower than for more complex chemical peptizers, these materials are used in larger quantities, relative to the amount of elastomer used. In addition to providing a milder form of peptization, these materials also have worthwhile mold release properties.

Besides the peptizers, a wide variety of materials are used as dispersing and processing agents to help the mastication process. Most of these materials differ from peptizers in that they do not react chemically with the polymer chains to break them. Examples of these agents are numerous; they include low-molecular-weight resins, fatty acids and their esters, and metal soaps. The resins are made to contain components that are compatible with elastomer aliphatic, aromatic, and naphthenic groups to improve homogeneity.

Fatty acids and their derivatives act as lubricants between polymer chains, causing easier flow during shearing when elastomers are being masticated. Most of these materials have only limited solubility in rubber compounds. They also reduce sticking of rubber compounds to processing equipment.

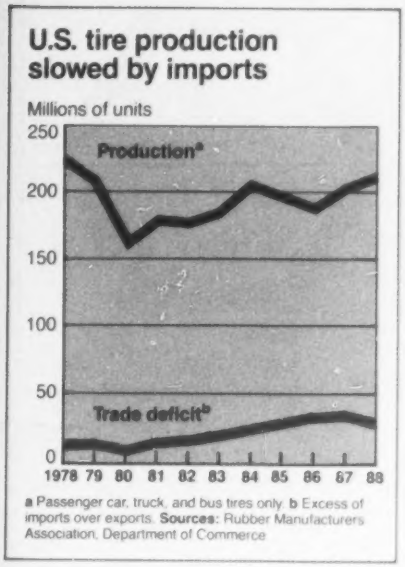
A host of other materials are used as softeners, plasticizers, and extenders of rubbers. Distinction between these materials and the dispersing and processing agents often is blurred because they do the same

Antidegradants make up more than half of rubber chemicals



Total 1987 production[c] = 382 million lb

a Antioxidants, antiozonants, stabilizers. b Includes activators and organic vulcanizing agents. c Excludes carbon black, sulfur, and fillers. Source: International Trade Commission


U.S. tire production slowed by imports
Millions of units
250
200
150
100
50
0
Production[a]
Trade deficit[b]
1978 79 80 81 82 83 84 85 86 87 88
a Passenger car, truck, and bus tires only. b Excess of imports over exports. Sources: Rubber Manufacturers Association, Department of Commerce

job. The exact material to be used will depend on the kind of elastomer going into the fabricated rubber product and on the properties wanted in that product.

Today a very large share of synthetic rubber that leaves the polymerization plant contains oil, especially the rubber destined to be made into tires and mechanical goods where color of the finished rubber product is black or doesn't matter. The combination of oils, carbon black, and polymer is called masterbatch. The oil undoubtedly has a softening effect on the elastomer. Later during processing, additional and different kinds of petroleum oils may be added to the compound. Many specifications are given for oils used in rubber processing, ranging from types of hydrocarbons present to viscosity. For example, oils with a higher content of aromatic hydrocarbons allow faster mixing of components of a rubber compound and faster cure rates during vulcanization. Aromatic oils, however, reduce both resiliency and good aging characteristics when compared with oils higher in paraffins.

Other materials used in lesser quantities in rubber processing as softeners and plasticizers are turpentine (useful because it contains significant quantities of terpenes that are oligomers of isoprene, a basic structure of natural and polyisoprene synthetic rubbers), pine tar, terpene resins, some synthetic resins (such as coumarone-indene and petroleum hydrocarbon resins), various esters, and natural fats and oils (other than those from pine trees). Small supply and high cost limit the use of many of these materials. Most will be used in low-production-volume products because they give specific advantages in cost/performance considerations. Properties of these materials cover a wide range and may be adversely affected by other components in a rubber compound.

Other materials are used in rubber compounding to provide certain properties in fabricated rubber products. The quantities of these materials used in rubber vary widely but, as a group, are much smaller than the accelerators and antidegradants. This miscellaneous group of rubber compound components includes bonding agents, flame retardants, colorants, and blowing agents.

For many fabricated rubber products, bonding agents are used to bind the elastomer to a reinforcing material for the products. The reinforcing materials are mostly metals and synthetic and glass fibers. A wide range of materials are used as bonding agents, depending on the composition of reinforcing material, form, and anticipated service.

Most steel wire used in reinforcing tires is first electroplated with brass. When the rubber compound contacts the brass as the tire is assembled, two competing chemical reactions occur at the interface of wire and rubber. In one reaction—a part of vulcanization—sulfur reacts with the elastomer. In the other, sulfur reacts with the copper in the brass. Good adhesion and long-term performance depend on high quality and uniform plating and upon avoidance of contaminants including water and nitrogen. Cobalt salts sometimes are used to improve the metal-elastomer adhesion.

Other methods of bonding elastomers to metals and textiles involve many different chemicals. Dipping the fiber in a mixture of resorcinol, formaldehyde, and latex (RFL) is basic to the bonding. Silanes may be used with glass fiber. Resorcinol and certain amines (such as hexamethyl tetramine as a methylene group do-

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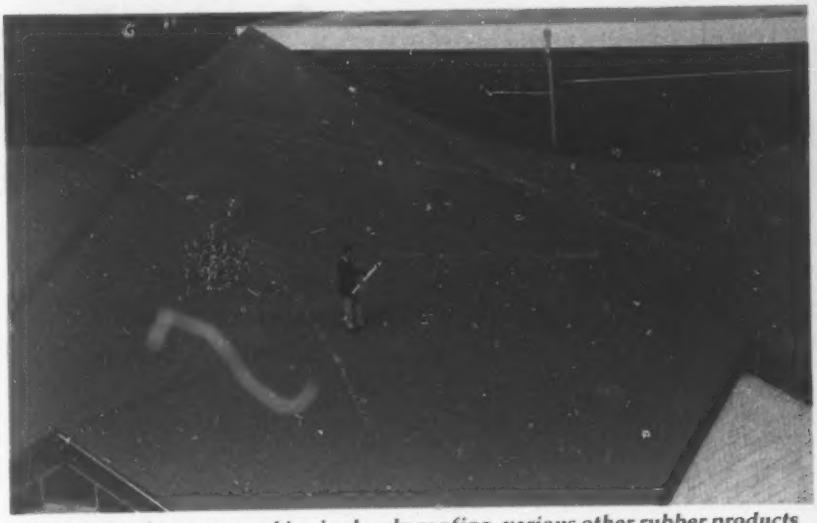
nor to polymerize the resorcinol) are added as bonding agents to the rubber compound, which is contacted with RFL-dipped reinforcements. Various complex isocyanates in organic solvents also may be applied by various physical methods to fibers, textiles, and metal parts, to bond them to elastomers.

Flame retardants are relatively high-cost chemicals added to rubber compounds when resistance to burning is considered to be a necessity. Examples are products such as carpet backing, underground conveyer belts, and single-ply roofing. Fillers, such as clays, aid flame retardancy. Frequently, antimony oxide and chlorinated paraffins or brominated aromatics are combined so that, if burning occurs, reaction products of antimony trihalides and steam dilute the oxygen around the burning part. Other flame retardants used less often are aluminum and magnesium hydroxides and ammonium phosphate. There are problems in using flame retardants in suitably high concentrations in that they may alter properties of the rubber product unacceptably.

Goodyear photo



Flame retardants are used in single-ply roofing, various other rubber products

Colorants are used in rubber compounds for a number of consumer products such as gloves, toys, bathing caps, and bottles. Cost, effect on properties, insolubility in the rubber compound, and compoundability are among factors that rubber processors consider in selecting colorants. For whites, titanium dioxide predominates. This chemical also is used with organic pigments to obtain pastel colors. Use of colorants often will require a change in the kind of other processing chemicals put into the compound to avoid staining or other deterrents to good coloring.

Blowing agents are used to produce the small quantities of foam or sponge rubber required in limited markets. Because most sponge rubber applicationss need closed-cell foams, blowing agents are selected to evolve gases, mostly nitrogen, by thermal decomposition. Among the commercially important blowing agents are complex azides, diazoaminobenzene, and some amines.

Still other chemicals are used for special purposes in compounding and fabricating rubber products, all developments of the long history of the consumption of elastomers in useful products. Further development of rubber-processing chemicals will be spurred by changing market needs. When demand becomes strong enough to render making rubber products for a market profitable, then production begins, bringing demand for processing chemicals along with it. □

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